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Synthesis and Performance Evaluation of Epoxy Resin–Modified Shape Memory Polyurethane Sealant

Reference

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ABSTRACT

To evaluate the effects of epoxy resin (EP) on different properties of shape memory polyurethane (SMPU) when used as a sealant with a tailored thermal transition temperature (T_t) for concrete pavement joints, the thermal and dynamic mechanical performances were discussed to determine the shape memory switching temperature of EP-modified SMPU, and then the compatibility between EP and SMPU, microscopic morphology, shape memory effect, and tensile property were also characterized. The results indicate that the tailored T_t of EP-modified SMPU can be used as the shape memory switching temperature to match its working temperature. EP and SMPU show considerable compatibility. EP inhibits the crystallization of soft phase content and increases the hard phase content. Both hydroxyl and epoxy groups participate in chemical reactions. The ring-opening reactions occur between fractional benzenes and SMPU. EP was successfully grafted onto the main chains of SMPU. Additionally, the peak load, tensile strength, and elongation at break of SMPU are first increased and then decreased when the EP content increase. An EP content of 10 % improves the tensile properties of SMPU, but excessive EP content leads to a decrease in the tensile strength and elongation at break. Finally, it is found that a small amount of EP has little influence on the shape memory properties of SMPU, but EP can improve the tensile properties of SMPU. EP-modified SMPU shows excellent shape memory effects. The prepared EP-modified SMPU with the specially tailored T_t can meet the requirements of practical engineering when used as a sealant for concrete pavement joints.

Keywords

epoxy resin, shape memory polyurethane, shape memory property, hard phase, soft phase

Introduction

Expansion joints are weak links in a concrete pavement system. Expansion joint failure is a leading cause of structural damage to concrete pavement [1]. Various types of sealants have been utilized to seal

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expansion joints in concrete pavements. These sealants are typically hot-poured, cold-poured, or preformed [1]. Among them, polyurethane (PU) is one of the most important sealant materials because of its excellent properties, such as good toughness, abrasion resistance, easy preparation, low price, etc [2]. However, synthesized PU usually shows a variety of properties that are due to differences in selected raw materials or their mixing ratios [3]. Recently, a novel two-component PU modified with hydroxyl that was named polydimethysiloxane was developed for use as a sealant of concrete pavement joints. This sealant showed satisfactory properties, such as tensile strength, cohesion strength, fatigue resistance, and aging resistance, when compared with conventional sealant, which led to an obvious decrease in expansion joint failure and pavement damages [4].

When PU material is specially designed, it can possess a shape memory effect because of its elastic network structure that consists of hard and soft phases [5]. The hard phase forms the net points that anchor the soft phase, keeping the original shape and playing a major role in the shape recovery process. The soft phase, on the other hand, maintains a temporary state after deformation [5]. This is similar to other thermally induced shape memory polymers (SMPs). It is known that SMPs are usually composed of two components on the molecular level, which includes the cross-links (fixed phase) to determine the permanent shape and the switching segments with transition temperature (T_t) to fix the temporary shape [6]. According to the nature of switching segments, SMPs are further subdivided into two categories, including SMPs with an amorphous switching segment, where T_t is the glass transition temperature (T_g) , and SMPs with a crystalline switching segment, where T_t is the melting temperature (T_m) [6].

More recently, SMP materials were utilized as sealant for expansion joints in concrete pavement or bridge decks [7]. Li and Xu [8] proposed that a SMP-based syntactic foam be used as a sealant for expansion joints after it had been programmed by two-dimensional (2-D) stress conditioning at a temperature above the T_{g} . Then, for practical applications, the programming with a one-stage 2-D stress condition was replaced by a sequential two-stage 1-D stress condition. To further simplify the programming process, 1-D tension programming was conducted at a temperature above the T_{g} followed by 1-D compression programming at a temperature below the T_{g} [9].

Among the above SMPs, the shape memory PU (SMPU), as a kind of thermally induced SMP, is seldom used as a sealant for expansion joints. However, SMPU not only shows the same advantages of conventional PU but also has unique properties. SMPU has become one of the most active SMP research fields because of its scientific and technological significance [10]. In general, SMPU is a block copolymer, which includes hard and soft segments. It often shows phase separation morphology because of the thermodynamic incompatibility between the two types of segments [10]. These block structure characteristics bring about obvious shape memory effects for SMPU. Hu et al. [11] revealed the phase-separated architecture of SMPU using dissipative particle dynamic simulations, which are of theoretical significance to the design of smart materials. Chiu et al. [12] used 2,6-pyridinedimethanol as a chain extender to prepare a novel SMPU. The T_g , maximum stress, and Young's modulus were improved with the increase in pyridinedimethanol content, but the elongation at break was reduced. Wu et al. [13] synthesized two types of PU using azobenzene as the hard phase, and test results showed that the two molecules exhibited trans-cis isomerization under ultraviolet irradiation.

However, SMPU also has limitations, such as small recovery force, low strength, poor crack resistance, weak durability, etc, that limit its application as an engineering material [14]. Therefore, SMPU is often reinforced by incorporation of different fillers, such as fibers, particles, nanotube, graphene, and so on, to improve its service properties [14]. Yong et al. [15] prepared reinforced SMPU using carbon nanotubes through conventional blending and crosslinking polymerization. The shape fixity ratio of reinforced SMPU was 92 %, and the shape recovery ratio was 95 %. Recently, Kim et al. [16] introduced nanographene particles into SMPU as a crosslinker and reinforcing filler and found that the shape fixity ratio, shape recovery ratio, yield strength, and T_g were all improved.

Additionally, the reactive liquid rubber, thermoplastic polymers, epoxy resin (EP), and silicone are also often used to modify PU to improve its service performance [17]. Among these, the EP is one of the most important thermosetting modifiers because of its high strength, good corrosion resistance, thermal stability, etc. In past decades, a considerable amount of work has been devoted to modifying PU using EP because of its excellent performance, which is generated by the interpenetrating polymer networks in the system [18].

Kalita and Karak [19] modified PU using a bisphenol-A-based EP, and the tensile strength, hardness, thermal stability, biodegradability, and gloss of the modified PU were improved with the increase in EP content. Zhan et al. [20] discussed the influence of EP on the water absorption, tensile strength, and stability of waterborne PU, and the effects of the two types of EP on the comprehensive properties of waterborne PU were also compared. Shen et al. [21] prepared EP-modified PU and found that the EP was successfully grafted onto the main chains of PU, which led to better thermal stability and mechanical properties.

It is noted that current EP-modified SMPUs still have some disadvantages; for example, the shape memory T_t , shape memory effect, and mechanical properties may not meet the requirements of practical engineering application [19]. Among them, a critical parameter for EP-modified SMPUs resides in its shape memory T_t . It is highly desirable that the shape memory T_t of EP-modified SMPU be tailored according to its practical application environment. The reason being that, when the working temperature is close to the T_t of EP-modified SMPUs, it may induce premature shape recovery in certain application environments [20]. Therefore, it is important to tailor the T_t of EP-modified SMPU to match its working temperature and actuate its shape memory behaviors. More importantly, a few studies focused on the EP's effect on the properties of SMPUs, although many studies were carried out on conventional EP-modified PU. It is not clear how the EP and its contents affect the shape memory T_t , shape memory effect, and mechanical properties of SMPU.

Therefore, the objective of this study is to develop a novel sealant to improve the service quality and extend the service life of expansion joints in concrete pavements. An EP-modified SMPU was synthesized to use as a sealant for expansion joints in concrete pavement with a tailored shape memory T_t range of 40°C–60°C. This is beneficial enough to trigger the viscoelastic relaxation process of the fixed shape for EP-modified SMPU, where the highest working temperature is around 70°C in China. Furthermore, the effects of EP on the shape memory T_t , shape memory effect, and mechanical properties of the EP-modified SMPU were discussed to provide a proper approach to improve the properties of SMPU using EP as a modifier and to determine suitable EP content to modify SMPU.

In this study, the EP-modified SMPU was first synthesized using the solution prepolymerization method. The thermal properties of EP-modified SMPU were tested to verify whether the tailored T_t was suitable as its working temperature by a differential scanning calorimeter (DSC). Then, a dynamic mechanical analyzer (DMA) was utilized to study the dynamic mechanical performance and further confirm the T_t of the prepared EP-modified SMPU. Fourier transform infrared spectroscopy (FTIR) tests were conducted to verify whether EP was successfully grafted onto the main chains of SMPU. Also, the field emission scanning electron microscopy (FESEM) and energy dispersive spectrometer (EDS) were used to observe the changes in microscopic morphology and chemical compositions of the SMPU before and after EP modification, respectively. Finally, the influence of EP contents on the shape memory effect and tensile properties of SMPU were discussed at the macro level. It is believed that this work may determine a suitable EP content that can be used to modify SMPU to improve its service properties.

Experimental

MATERIALS AND SYNTHESIS OF SAMPLES

Poly(1,4-butylene adipate glycol) (PBAG; Xuchuan Chemical Co., Ltd., Suzhou, China; $M_n = 2,000$), 2,4-tolylene diisocyanate (TDI; TCI Chemical Co., Ltd., Shanghai, China), 1,4-Butanediol (BDO; Sinopharm Chemical Reagent Co., Ltd., Shanghai, China), and EP (Nantong Xingchen Synthetic Material, Co., Ltd., Nantong, China) were used as raw materials to synthesize EP-modified SMPU in this study.

The synthesis was carried out in a 250-ml and four-neck round-bottomed flask, which was equipped with a thermometer,

a mechanical stirrer, and nitrogen inlet and outlet tubes. First, the calculated amount of PBAG was put into the flask. Then, the temperature was slowly elevated to 120°C and maintained for 1.5 h for vacuum dehydration, wherein the vacuum degree was more than 0.095 MPa.

Second, when the temperature had been slowly dropped to 80°C, the calculated amount of TDI was added. The reaction took place under the protection of nitrogen, and the temperature was stabilized at 80°C for 2 h to obtain the prepolymer of SMPU. Third, when the temperature was further dropped to 70°C, the calculated amount of EP was added to the prepolymer. Then, the required amounts of BDO were added, dropwise. The mixtures were blended quickly by mechanical stirring for 30 min at 70°C to achieve a complete reaction.

Finally, when the reaction was complete, the EP-modified SMPU was immediately injected into a polytetrafluoroethylene mold where the homogeneous mixture was cooled to room temperature and cured. Thus, the sample of EP-modified SMPU was obtained after demolding. This sample was then subjected to different experiments to test its various properties. The prepared samples were marked as SMPU, 10 % EP/SMPU, and 20 % EP/SMPU, which represent the ratios of EP to SMPU prepolymer as 0, 10, and 20 % by weight, respectively.

CHARACTERIZATION METHODS

DSC Test

A 204 F1 Phoenix DSC (NETZSCH, Selb, Germany) was used to analyze the effects of the EP content on the T_t of the SMPU under a nitrogen atmosphere. Approximately 10 mg of the sample were heated from -20° C to 140°C at a heating rate of 10°C/min. Subsequently, the sample temperature was dropped to -20° C at a cooling rate of 20°C/min. Once again, the sample was heated to 140°C at a rate of 10°C/min.

DMA Test

A Q800 DMA (TA Instruments, New Castle, DE) was utilized to study the thermodynamic properties of the prepared SMPU samples. The samples were heated from -50° C to 100° C at a heating rate of 5°C/min with a frequency of 1 Hz.

FIRT Test

In order to verify whether SMPU was synthesized or the EP was successfully grafted onto the main chains of the SMPU, FTIR analysis was performed using a Bruker Vector 22 FTIR spectrometer (Bruker, Billerica, MA). Each sample was prepared by casting film onto a potassium bromide (KBr) thin plate. The spectra were collected in the wave number range from 4,000 to 500 cm⁻¹ at a resolution of 4 cm⁻¹.

FESEM and EDS Test

A JSM-7600F Schottky FESEM (JEOL Ltd., Tokyo, Japan) equipped with an EDS was used to observe microscopic morphology characteristics and identify chemical compositions of pure and EP-modified SMPU, respectively. Samples were first fixed on an aluminum sample stub and sputtered with gold under vacuum conditions. Then, the sample chamber was opened in order to place the samples. Finally, the morphologies of the samples were observed using FESEM, and chemical compositions were detected using the EDS.

Shape Memory Effect Test

To evaluate whether the addition of EP affected the shape memory effect of the SMPU, the prepared sample was first machined into dog bone-shaped specimens. Their middle central length was recorded as l_0 , and both ends of the specimens were clamped in a rhombus-shaped truss fixture for uniaxial stretching, as shown in Fig. 1. Then, the truss fixture with the prepared specimen was placed in a heating chamber at $(T_t + 10)^{\circ}$ C for 20 min to make the temperature distribute uniformly in the specimen. Subsequently, a constant load of 49 N was applied on the truss fixture to stretch the specimen at $(T_t + 10)^{\circ}$ C for 30 min to stabilize the deformation. After that, the heating was first stopped, the chamber was kept closed, and the system was allowed to naturally cool to room temperature while maintaining the applied load constant. The tagged middle central length was recorded as l_1 . After removing the load, the specimen was placed on a slab at room temperature for 24 h, and the tagged middle central length was recorded as l_2 . Finally, the specimen was placed back in the chamber to recover freely for 30 min at $(T_t + 10)$ °C. The tagged middle central length was recorded as l_3 . The shape fixity ratio (R_f) and shape recovery ratio (R_r) were expressed as follows [22]:

$$R_f = (l_2 - l_0) / (l_1 - l_0) \times 100\%$$

$$R_r = (l_2 - l_3) / (l_2 - l_0) \times 100 \%$$
⁽²⁾

FIG. 1 Stretching method of the SMPU specimen in a heating chamber.



Tension Test

Dog bone specimens with middle distances of 40 mm were stretched by an ETM504C electronic universal testing machine (Wance, Shanghai, China) to study the influence of EP content on the tensile properties of the SMPU. The tensile properties, such as tensile strength and elongation at break, were tested at room temperature with a loading rate of 10 mm/min. Three effective specimens were tested for each group.

Results and Discussion

THERMAL PROPERTIES

Thermally responsive SMP materials can change or restore their shape at a specific temperature range. The thermal transition of SMPU is essential to determine the test conditions and understand the shape memory behaviors. The DSC tests are conducted to analyze effects of the EP on the thermal properties of SMPU. The test results are shown in **Fig. 2**.

As shown in **Fig. 2**, the obvious step-shape decreasing curves are not observed, thus it is difficult to confirm the T_g of EPmodified SMPU. However, the endothermic peaks are observed in a temperature range of 40°C–60°C, which reveals the T_m s of crystallites in soft segments. The T_m s values of the SMPU samples are 56.9°C, 51.6°C, and 46.9°C as the EP content is increased from 0 to 20 %, respectively. Also, it is found that there is only one endothermic peak on the DSC curves, which suggests considerable compatibility between the EP and SMPU. Furthermore, the DSC curve peak shows a shift to the low temperature, indicating that the T_m values of the SMPUs are decreased gradually. This is because the hard phase fraction in the SMPU becomes relatively larger as the EP content is increased. It inhibits the crystallization



of the soft phase, leading to the decrease in crystallinity of SMPU [23]. As a result, the T_m of SMPU is lowered as the EP content increases.

Additionally, the T_m of the hard phase is usually higher than this temperature range, so the DSC curve peaks in **Fig. 2** are attributed to the endothermic reactions of the soft phase [24]. Because the shape memory behavior is usually generated by the entropy elastic behavior of the rubbery soft phase, the T_m of soft segments is generally regarded as T_t of the SMPU to actuate the shape memory actions of SMPU [24]. Therefore, DSC thermograms determine the T_m of crystalline soft segment as the shape memory switching temperature of prepared SMPU in this study [25].

DYNAMIC MECHANICAL PERFORMANCE

DMA is used to investigate the influence of the EP on the dynamic mechanical properties of SMPU. It is known that the loss and storage modulus can be used to study the shape memory properties and rigidity of SMPU. The loss tangent (tan δ) is defined by the ratio of loss modulus (E'') to storage modulus (E'), which shows such information as damping capability and T_t [26]. tan δ test results of the SMPU samples with different contents of the EP are shown in **Fig. 3**.

Fig. 3 shows the dynamic mechanical properties of the SMPU with different EP contents. It is seen that the tan δ curve peaks are present in the temperature range of SMPU thermal transition region. Furthermore, the temperatures to which the tan δ curve peaks correspond are the T_t values of the SMPU with different EP contents. The T_t values of the SMPU, 10 % EP/SMPU, and 20 % EP/SMPU are 48.2°C, 44.3°C, and 39.6°C, respectively. It is also noted that the tan δ curves show a shift to low temperature with the increase in EP content, indicating that the T_t of the SMPU is lower. A possible reason is that the addition of EP leads



to a decrease in the crystallographic capacity of the soft phase in the SMPU [26].

At the same time, the peaks of tan δ curves become wider with the increase in EP content, suggesting that the length distribution of molecular chains is becoming more nonuniform. This is because the phase proportion, which can cause thermal transition, is reduced with the increase in EP content. Consequently, this causes the length distribution of molecular chains in the SMPU to be more nonuniform [26]. Test results of T_t using DMA are consistent with those results measured by DSC, although there is a deviation because of different testing principles and measuring techniques between the two experimental instruments. DSC measures the endothermic processes while DMA reflects the dynamics of the sample. However, in general, the results of these two tests can determine the T_t values of EP-modified SMPU to study its shape memory properties. This is consist with the test results in the previous study [25].

FTIR SPECTRUM ANALYSIS

FTIR tests are used to confirm whether the EP is successfully grafted onto the main chains of the SMPU. Test results of the FTIR spectra are presented in **Fig. 4**.

From Fig. 4a, it is clearly observed that the band at 3,500 cm⁻¹ is due to the vibration absorption of hydroxyl (–OH) in the main chains of the EP. The band at 1,295 cm⁻¹ is attributed to quaternary carbon atoms in the EP. The band at 913 cm⁻¹ is assigned to the epoxy group, and the band at 826 cm⁻¹ shows the presence of benzene skeletons in the EP. All of these are characteristic bands of EPs [27].

As shown in **Fig. 4b–d**, the band at 3,338 cm⁻¹ is caused by the stretching vibration of amidogen (–NH). The band at 1,725 cm⁻¹ is due to the stretching vibration of carbonyl (C=O) in the urethane



group. The band at 1,533 cm⁻¹ shows the existence of carbamate, which is generated by the reaction between –OH in PBAG and isocyanate (–NCO) in TDI [27]. The band at 1,225 cm⁻¹ is caused by the stretching vibration of the carbon-oxygen bond in the ester group. The presence of these characteristic bands indicates that PBAG reacts with TDI and BDO to generate SMPU [2].

Compared with Fig. 4a and b, it is seen in Fig. 4c and d that the characteristic bands at 3,500 cm⁻¹ of –OH in EP and 913 cm⁻¹ of epoxy groups disappear. This suggests that both –OH and epoxy groups participate in chemical reactions during the preparation of EP-modified SMPU. They were consumed completely in this reaction [26]. A weak stretching peak of benzene-ring skeletons appears at the band of 1,503 cm⁻¹. The characteristic band at 1,290 cm⁻¹ shows the presence of quaternary carbon atoms. The characteristic peak of benzene rings at the band of 826 cm⁻¹ is still present, though its intensity is weak. A possible reason for this is that ring-opening reactions between fractional benzenes and SMPU occurred [26]. All these indicate that the EP was successfully grafted onto the molecular chains of SMPU.

MORPHOLOGY AND CHEMICAL COMPOSITIONS

FESEM and EDS are used to analyze the changes in morphology and elemental composition between pure and EP-modified SMPU samples and to confirm whether the EP is compatible with SMPU. The SEM images and elemental compositions of SMPU with different EP contents are illustrated in **Figs. 5–7**.

Figs. 5a, 6a, and 7a show SEM images of SMPU with different EP contents, wherein the bright region consists of hard segments and the dark region is composed of soft segments. As the EP content increases, the dark region becomes smaller and the bright region becomes larger, indicating that the hard segments in SMPU increased.

FIG. 5 (a) SEM image and (b) EDS spectrogram of SMPU sample.



FIG. 6 (a) SEM image and (b) EDS spectrogram of 10 %EP/SMPU sample.



FIG. 7 (a) SEM image and (b) EDS spectrogram of 20 % EP/SMPU sample.



This is because the benzene rings in epoxy chains of the EP react with polar groups, such as –NH, C=O, in the SMPU. As a result, the interaction force and compatibility between EP and SMPU are enhanced, and the hard phase fraction in SMPU is increased [27]. Additionally, compared with Fig. 5a, the morphology of samples in Figs. 6a and 7a becomes smoother and more uniform without obvious phase separation phenomenon, suggesting that EP and SMPU are fully integrated. The dispersed phase was uniformly distributed in the continuous phase of matrix.

From **Figs. 5b**, **6b**, and **7b**, only carbon (C) and oxygen (O) elements are detected in pure and EP-modified SMPU. Hydrogen may also exist, but its molecular weight is too small to be detected. It is found that there is a little difference in the element content of C and O in each sample. However, as the EP content is increased, the C content is slightly increased while the O content is decreased. This may be due to the fact that the ratio of C to O in EP is larger than that in SMPU.

SHAPE MEMORY PROPERTY

Shape memory effect tests are performed to investigate the effects of EP content on the shape memory properties of SMPU. The comparative photographs of the pure SMPU specimens at different test steps, as a representative, are shown in **Fig. 8**. Test results are presented in **Table 1**.

 TABLE 1
 Test results of shape memory effect of the SMPU specimens with different EP contents.

EP Content (%)	l_0 (mm)	l_1 (mm)	$l_2 \text{ (mm)}$	<i>l</i> ₃ (mm)	R_f (%)	R_r (%)
0	40.0	42.7	42.7	40.0	100	100
10	40.0	43.0	43.0	40.0	100	100
20	40.0	42.0	41.9	40.0	95	100

Fig. 8a shows the original specimen before stretching, and the stretched specimen in **Fig. 8b** is obtained after the original specimen is unloaded. When the temperature rises above the T_t of the SMPU and is maintained for 30 min, the stretched specimen is freely recovered, as shown in **Fig. 8c**. It can be seen that the recovered specimen of the pure SMPU is the same length as the original one, as shown in **Table 1**. As a result, both R_f and R_r of the pure SMPU are 100 %. Similarly, it is seen from **Table 1** that both R_f and R_r of the 10 % EP/SMPU specimen are 100 % while the R_f of the 20 % EP/SMPU specimen is 95 % and its R_r is 100 %.

It is found that the R_r s of both the pure and EP-modified SMPU are larger than 95 %, and their R_f s are 100 %, indicating they have good shape memory properties. The slight decrease in the R_f for the 20 % EP/SMPU specimen may be due to the larger spring-back of the excessive EP component after unloading.



FIG. 8 (a) Original specimen, (b) stretched specimen after unloading, and (c) specimen after recovery of pure SMPU.

However, EP reinforces the hard segments of the SMPU, maintaining the 100 % R_r , even in the 20 % EP/SMPU specimen.

TENSILE PROPERTY

Tensile property is one of the most important properties of SMPU because it affects its scope of application. Tension test results are given in **Fig. 9** and **Table 2**.

From Fig. 9 and Table 2, it can be seen that the tensile strength and elongation at break of the SMPU first increases and then decreases based on an increase in the EP content, and they both reach the maximum when the content of the EP is 10 %. The reason for the increase in tensile strength and elongation at break is that when a small amount of EP is added into the SMPU, crosslinking reactions occur between –OH in the EP and –NCO in the SMPU [28]. Also, the ring-opening reactions of the fractional EP occur in the system so that the interpenetrating network structure is formed and the tensile properties of the SMPU are improved [29].

However, -NCO in the SMPU reacts completely with -OH in the EP when the EP content is further increased, according to the test results of FTIR. The excessive EP that is not involved in the reaction is left in the system. The modified SMPU becomes

 TABLE 2
 Tensile properties of SMPU specimens with different EP contents.

EP Content (%)	Peak Load (N)	Tensile Strength (MPa)	Elongation at Break (%)
0	973.22	17.36	13.91
10	1,002.00	18.56	17.94
20	513.69	9.51	11.65

FIG. 9	Tensile stress-strain responses of the SMPU specimens wi	ith
	different EP contents.	



more brittle because of the low toughness and poor crack resistance of the EP, leading to a decrease in tensile strength and elongation at break [29]. It is concluded that a suitable EP content is needed to improve the mechanical properties of the SMPU.

Conclusions

In this study, EP-modified SMPU samples were first synthesized, and their different properties were characterized. Based on the comprehensive property characterization and analysis, the following conclusions are obtained:

- (1) The tailored T_t of EP-modified SMPU can be used as the shape memory switching temperature to actuate its shape memory behaviors, which match its working temperature when used as a sealant of pavement joints. Test results indicate a considerable compatibility between EP and SMPU. The EP inhibits the crystallization of the soft phase and increases the hard phase content in SMPU.
- (2) Both –OH and epoxy groups participate in chemical reactions during the preparation of EP-modified SMPU. The ring-opening reactions occur between fractional benzenes and the SMPU. FTIR spectra of EP-modified SMPU indicate that EP is successfully grafted onto the main chains in the SMPU.
- (3) The addition of EP increases the hard phase content of the SMPU, and the crack surface becomes smoother and more uniform without obvious phase separation phenomenon. Only C and O are detected in the pure and EP-modified SMPU. The EP has a little influence on the elemental compositions of the SMPU.
- (4) The peak load, tensile strength, and elongation at break of the SMPU are increased first and then decreased with the increase in EP content. A suitable EP content of 10 % is confirmed to improve the tensile properties of the SMPU, and the excessive addition of EP causes SMPU to become more brittle, leading to the decrease in tensile strength and elongation at break of the SMPU.
- (5) A small amount of EP has little influence on the shape memory property of SMPU, but the EP can improve its tensile performance. The EP-modified SMPU still shows excellent shape memory effect. The EP-modified SMPU with the specially tailored T_t can meet the requirements of practical engineering applications when used as a sealant for concrete pavement joints.

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